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## **The application of retrospective luminescence dosimetry in areas affected by fallout from the Semipalatinsk Nuclear Test Site: an evaluation of potential**

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## **Abstract**

Luminescence retrospective dosimetry techniques have been applied with ceramic bricks to determine the cumulative external gamma dose due to fallout, primarily from the 1949 test, in populated regions lying NE of the Semipalatinsk Nuclear Test Site in Altai, Russia, and the Semipalatinsk region, Kazakhstan. As part of a pilot study, nine settlements were examined, three within the regions of highest predicted dose (Dolon' in Kazakshstan; Laptev Log and Leshoz Topolinskiy in Russia) and the remainder of lower predicted dose (Akkol', Bol'shaya Vladimrovka, Kanonerka and Izvestka in Kazakshstan; Rubtsovsk and Kuria in Russia) within the lateral regions of the fallout trace due to the 1949 test. The settlement of Kainar, mainly affected by the 24 September 1951 nuclear test, was also examined. The bricks from this region were found to be generally suitable for use with the luminescence method. Estimates of cumulative absorbed dose in air due to fallout for Dolon' and Kanonerka in Kazakshstan and Leshoz Topolinskiy were  $475 \pm 110$  mGy,  $240 \pm 60$  mGy and  $230 \pm 70$  mGy, respectively. The result obtained in Dolon' village is in agreement with published calculated estimates of dose normalized to  $^{137}\text{Cs}$  concentration in soil. At all the other locations (except Kainar) the experimental values of cumulative absorbed dose obtained indicated no significant dose due to fallout that could be detected within a margin of about 25 mGy. The results demonstrate the potential suitability of the luminescence method to map variations in cumulative dose within the relatively narrow corridor of fallout distribution from the 1949 test. Such work is needed to provide the basis for accurate dose reconstruction in settlements since the predominance of short-lived radionuclides in the fallout and a high degree of heterogeneity in the distribution of fallout are problematic for the application of conventional dosimetry techniques.

**Keywords:** Semipalatinsk Nuclear Test Site; retrospective luminescence dosimetry; fallout; environmental; radiation dose

## **Introduction**

It is now acknowledged that there exists a major dosimetry problem requiring further investigation (Simon et al. 2003; Simon and Bouville 2002; Bouville et al. 2002; Gilbert et al. 2002) within the populated regions adjacent to the Semipalatinsk Nuclear Test Site (SNTS). Substantial releases of radioactive fallout from, in particular, the 1949 tests delivered significant doses to inhabitants of certain areas adjacent to the Test Site, notably those in the Altai region, Russia, and the Semipalatinsk region, Kazakhstan. The application of conventional dose reconstruction methods is problematic, largely due to the predominance of short-lived radionuclides in the fallout, which are now absent in contemporary soil assays. Although published dose estimates for this region are available (Shoikhet et al. 1998; Stepanenko et al. 1994; Stepanov et al. 2002; Gordeev et al. 2002; Gusev et al. 1997; Stepanenko 1989), they are based on calculations that employ a very limited quantity of historical data comprising exposure rate and radionuclide concentration measurements performed following the tests and available parameters related to the explosion. In addition, a high degree of heterogeneity in the distribution of fallout is likely to have occurred. For these reasons alternative approaches to the retrospective assessment of radiation dose are needed, and in this work we report on an investigation of the potential of the experimental method of luminescence retrospective dosimetry to contribute to dose reconstruction in this region.

The fallout from the nuclear test performed on 29 August 1949, considered to be the main source of radiation dose, moved rapidly from the SNTS in a NEE direction (Fig. 1) due to strong winds that developed on the day of the test. This led to a relatively narrow corridor of affected territory within which populated settlements were located (Shoikhet et al. 1998). The method of luminescence retrospective dosimetry (ICRU 2002) was applied in this work to determine the cumulative external gamma dose due to fallout by testing fired clay bricks (FCBs) taken from buildings that were constructed before the tests. The methodology applied is a development of that used in populated

areas in Ukraine and Russia affected by fallout from the Chernobyl Nuclear Power Plant (Bailiff et al. 2003). The overall aim was to survey, assess and test the use of the method in populated areas affected by fallout from the SNTS that are now located in Kazakhstan and the Russian Federation. Takada et al. (1997; 2002) reported the outcome of preliminary luminescence work in several settlements downwind of the 1949 tests in Kazakhstan. This work included the village of Dolon' and other settlements near the SNTS which are currently the focus of considerable epidemiological interest (see, for example, Shoikhet et al. 1999) because of the high levels of cumulative dose predicted by calculation. Also of note is the preliminary work by Ivannikov et al. (2002) who, using electron paramagnetic resonance (EPR) spectroscopy with samples of human tooth enamel taken from residents living in the vicinity of the Semipalatinsk nuclear test site, found a significant difference between EPR dose estimates and previously reported luminescence (Takada et al. 2002) and calculated dose estimates (Tsyb et al. 1990; Stepanov et al. 2002; Gordeev et al. 2002; Gusev et al. 1997) for the village of Dolon'. The EPR dose estimates obtained by Ivannikov and co-workers for three samples of teeth from inhabitants of Dolon' village were about 6-7 times lower than luminescence data and calculated values based on historical exposure rate measurements. However, as noted elsewhere (ICRU 2002) and worth stressing here again, EPR dose estimates are based on measurements with materials taken from individuals, whereas in the case of luminescence the relevant materials are taken from the environment and it therefore has a different role in dose reconstruction. In the context of this project the role of interest is the validation of the calculated estimates of cumulative dose in populated areas referred to above.

### **Summary of methodology**

The experimental quantity determined using the luminescence method is the cumulative absorbed dose in brick since its manufacture,  $D_T$ . The quantity of interest in retrospective dosimetry is the

cumulative absorbed dose since the onset of the delivery of fallout in the vicinity of a sampled building,  $D_X$  (Bailiff 1997). The latter is obtained by calculating the difference between  $D_T$  and the cumulative natural background dose,  $D_{BG}$ :

$$D_X = D_T - D_{BG} \quad (1)$$

In this work  $D_T$  was determined by applying established luminescence techniques with crystals of quartz extracted from the ceramic (brick), and  $D_{BG}$  was determined, in common with previous studies (e.g., Bailiff et al. 2003), by calculating the product of the component dose-rates due to natural sources of radiation and the known age,  $A$  years, of the ceramic sample:

$$D_{BG} = A(b\dot{D}_\beta + g\dot{D}_\gamma + \dot{D}_{cos}) \quad (2)$$

The terms  $b\dot{D}_\beta$  and  $g\dot{D}_\gamma$  are the annual beta and gamma ray dose arising from natural sources of radiation for the quartz grains extracted for luminescence measurements. By selection of grains of sufficient size and the application of chemical etching treatments the alpha dose can be neglected (ICRU 2002). The constants  $b$  and  $g$  are related to attenuation effects and irradiation geometry respectively, and  $\dot{D}_{cos}$  is the annual dose due to cosmic-rays. The gamma ray component of  $D_{BG}$  can also be obtained more directly using dosimeters to measure *in situ* the combined gamma and cosmic ray dose-rate,  $\dot{D}_{cap}$ . In this case Eq. 2 becomes

$$D_{BG} = A(b\dot{D}_\beta + \dot{D}_{cap}), \quad (3)$$

where it is assumed that the section of the building and its local environment have not changed significantly since construction of the building and that the concentration of extant artificial radionuclides is sufficiently small to make a negligible contribution to the dose recorded using the

dosimeter. As discussed further below,  $\dot{D}_{\text{cap}}$  is expected to vary with position in the wall, in particular with depth.

Providing there is a measurable difference between  $D_T$  and  $D_{BG}$ , the value of  $D_X$  is converted to absorbed dose in air at a reference location,  ${}_{RL}D_X$ , to allow comparisons with estimates of cumulative dose arrived at by modelling calculations where,

$${}_{RL}D_X = C_{RL} \cdot D_X \quad (4)$$

The conversion factor  $C_{RL}$ , defined as the inverse of the ratio of the absorbed dose in brick to the air kerma at the reference location, has been calculated for a range of energies and geometries on the basis of Monte Carlo (MC) simulations (ICRU 2002; Bailiff et al. 2003; Jacob et al. 2000).

When determining  $D_T$  and calculating  $C_{RL}$ , certain assumptions are made concerning the time-averaged source energy and source configuration (Bailiff et al. 2003). The mass energy absorption coefficient for quartz (and other silicates) increases substantially for photon energies below 100 keV. An assessment of the potential contribution to the absorbed dose  $D_T$  by low energy photons is required because experimental determinations of dose by luminescence are performed using  ${}^{90}\text{Sr}/{}^{90}\text{Y}$  beta sources that are calibrated against a secondary standard  ${}^{137}\text{Cs}$  photon source. On the basis of available information concerning the fallout inventory from the tests (Izrael and Stukin 1967), the time-averaged mean source energy for fallout from the 1949 Semipalatinsk tests is estimated to lie in the range 500-800 keV, and hence the proportion of absorbed dose arising from photons of energy  $< 100$  keV is likely to be small. Also, no further correction to the calculation of  $D_{BG}$  is required since less than 5% of the total energy emitted by the naturally occurring radionuclides is carried by photons of energy less than 100 keV (Aitken 1985).

The measurement of depth-dose profiles in brick potentially provides the opportunity to test experimentally assumptions concerning the source energy and configuration, although the profile

cannot be used to unambiguously reconstruct the source energy because it is a function of both energy and geometry (Meckbach et al. 1996). As the source energy decreases the profile becomes steeper for a given source geometry, reflecting the shorter mean free path of the photons; however MC simulations also predict that a relatively steeper profile is obtained if sources of a given energy are located entirely on the wall surface rather than in the ground. In the case of a source energy of 662 keV, for example, the 'half-depth' reduces from  $\sim 6$  cm to  $\sim 2.5$  cm for ground and wall source configurations, respectively (Fig. 2; ICRU 2002).

### **Study sites**

On the basis of the published calculations of cumulative dose for the 29 August 1949 nuclear test (Logachev 1997; Shoikhet et al. 1998), we initially sought samples in settlements located at two distances from the detonation point that lay close to the central axis of the plume (i.e. highest dose) and settlements at comparable distances but lying on transects orthogonal to the main axis in regions of significantly lower fallout (Fig. 1). However, brick was not a commonly used building material during the 20<sup>th</sup> century, particularly in Kazakhstan. Work performed by scientists based in Kazakhstan (Almaty and Semipalatinsk) and Altai, Russia (Barnaul) identified 14 potentially suitable settlements within the regions of highest predicted dose and those of lowest predicted dose within the lateral regions of the fallout trace arising from the 1949 tests. However, one of the settlements (Kainar) sampled was mainly affected by the 24 September 1951 nuclear test (Logachev 1997). Ten of the locations (Fig. 1) were sampled during fieldwork expeditions conducted separately in Kazakhstan and Russia in 1999 and 2000, respectively. Local building authority documentation and historical records were examined to establish the date of construction and local residents were also consulted concerning alterations to the buildings. A summary of the details of the buildings and the sampled locations is given in Table 1. Records indicated that eight of the buildings were constructed between 1900 and 1930, that one was built in 1947 (Rubtsovsk), and the sampled chimney on the building in Kainar was reported to have been constructed before



1949. Figs 3a-e show the physical context and nature of three of the buildings sampled, two of which were of particular interest because of their location in the region of highest fallout (Dolon' and Leshoz Topolinskiy).

In three buildings (Dolon', Kanonerka and Bol'shaya Vladimirovka) it was possible to obtain brick from the interior of the building and perform an independent check of the age. As mentioned above, brick buildings are scarce in the region of Kazakhstan investigated and one of the difficulties encountered was their use as a source of recycled building materials. In one case (Kannerka) the building had been demolished within one year of sampling.

The brick walls of the buildings sampled in Dolon', Bol'shaya Vladimirovka and Laptev Log were rendered with mortar (average thickness given in Table 1). Since the render is located on the surface of the wall, the dosimetry of the immediate sub-surface is lost since mortar is not suitable for dose determinations at these levels using current luminescence dosimetry techniques. A critical aspect in terms of interpretation of the luminescence results is whether the render was added in one or more layers between the delivery of fallout and sampling. The available building history for Dolon' and Bol'shaya Vladimirovka suggests that the render was in place before 1949, but that, in the case of Laptev Log, currently available information suggests that it had been applied in *ca* 1988. Consequently the depth of brick extracted from rendered walls was taken in this study to be the depth below the surface that was present during the delivery of fallout and assumed to be in place for at least 1 year following the tests during which at least 90% of the cumulative dose to the present day was delivered (Shoikhet et al. 1998).

Samples of brick were obtained either by extracting whole bricks or by using a diamond-faced corer attached to an electric drill that enabled 50 mm diameter brick cores to be taken from the surface to

the full depth of the brick depending on its orientation in the wall (generally 12 cm, but in some cases > 30 cm). The speed of drilling was regulated to moderate the surface temperature of the core and, once extracted, the cores were double bagged in heavy gauge black polythene. The standard height of sampling for ‘ground level’ samples was ~1 m above the ground immediately adjacent to the sampled wall. In the cases of the mill in Leshoz Topolinskiy and the former church in Kuria it was also possible to obtain samples at elevated heights (Table 1). Whole bricks were cut into four main sections, retaining the full depth of the brick in each case to allow distribution to different laboratories; and in some cases they were subsequently cut into narrower sections. As illustrated in Fig. 4, samples were coded by location number (e.g. 73-1) with the addition of a section number (e.g. 73-1-1) and any further dissection, if performed (e.g. 73-1-1-1).

The gamma dose-rate was measured with an Automess meter 6150-AD-1 and a GM probe type 18<sup>1</sup> and recorded at the sampling location, and also at intervals along a line orthogonal to the sampled wall extending up to ~40 m at heights of ~20 cm and ~1 m above the ground surface. This was done to investigate variations in the natural gamma radiation field and to detect the presence of artificial radionuclide activity. At all the sites examined the dose-rates measured were consistent with levels expected from the presence of natural radionuclides (Table 2) and extant levels of <sup>137</sup>Cs detected at some of the sites. Al<sub>2</sub>O<sub>3</sub>:C luminescent dosimeters (Akselrod et al. 1990) were deposited in holes drilled adjacent to the location of the brick samples to enable the measurement of the absorbed gamma dose in the wall by means of TL measurements in the laboratory. Details of the thickness of the sampled wall and dimensions of the building and topography of the adjacent ground were recorded.

### *Soil samples*

Samples of soil from ground adjacent to the sampled buildings were taken using a standard soil corer to determine the quantity of lithogenic ( $^{238}\text{U}$ ,  $^{232}\text{Th}$  series and  $^{40}\text{K}$ ) and any extant artificial radionuclides such as  $^{137}\text{Cs}$ . Generally cores to a depth of 5-20 cm were obtained, although longer cores were taken at some locations depending on soil type. The soil sampling was performed where the ground was judged not to have been disturbed. Where this was not possible samples of soil were taken from undisturbed ground at the limits of the settlement. Cores were cut into 5 cm length sections and individually bagged to enable a depth-activity profile to be produced (Isvestka, Kainar, Rubtsovsk, Laptev Log, Leshoz Topolinskiy, Kuria).

### **Experimental**

The required brick sections were cut using a water-lubricated diamond blade to produce a series of slices of increasing depth ranges from the front surface of the wall. The central depths were located at 10, 20, 40, 60 and 100 mm and the thickness of the slices was generally ~4 mm in the sub-surface layers, increasing to ~10 mm at greater depths if a higher yield of quartz grains was required. In cases where a significant reduction of  $D_T$  with depth was detected, measurements were performed to obtain a depth-dose profile with finer depth resolution (Locs 71, Dolon'; 73, Kanonerka; 82/83, Leshoz Topolinskiy).

Clay and crystalline mineral grains were extracted from the slices using mechanical crushing and sieving procedures. Following the quartz inclusion technique developed for archaeological dating applications (Aitken 1985; 1998), grains of selected size ranges within the overall range 90-200  $\mu\text{m}$  were subjected to an hydrofluoric acid (40%) etching treatment to remove the outer layer of the

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<sup>1</sup> available from Automess GmbH, Daimlerstrasse 27, D-68526 Ladenburg, Germany

grains and to isolate the quartz fraction by removal of other silicate minerals. Checks for the presence of residual feldspars made with randomly selected aliquots by testing for the presence of infra-red stimulated luminescence indicated that the luminescence detected was primarily from quartz. Heavy liquid separation procedures were also used to further purify the quartz fraction if required. By removing the outer layer of the grains, the absorbed dose contribution from alpha particles emitted by radionuclides in the brick fabric ( $^{238}\text{U}$ ,  $^{232}\text{Th}$  and progeny) is reduced to a negligible level (Aitken 1985).

Two luminescence techniques were applied to determine  $D_T$  for quartz grains. They are based on different luminescence mechanisms associated with quartz i) the 210 °C thermoluminescence (TL) peak (Bailiff and Petrov 1999; Göksu et al. 2001) and ii) optically stimulated luminescence (OSL; stimulation wavelength range ~450 - ~550 nm; Godfrey-Smith and Haskell 1993; Boetter-Jensen et al. 2000), the experimental procedures for which are discussed in more detail by Bailiff et al. (2000). Techniques based on different luminescence mechanisms are used to obtain a cross check of the reliability of dose evaluations. The measurements were performed by four groups (Durham (DUR), GSF Neuherberg (GSF), Helsinki (HEL), and MRRC Obninsk (MRRC)) in three laboratories (DUR, GSF and HEL) using semi-automated readers of similar type manufactured by the Risø National Laboratory<sup>2</sup>. The beta radiation sources used to administer laboratory doses were calibrated against a common secondary standard  $^{60}\text{Co}$  photon source at the GSF Laboratory for luminescent sample of specific type, thickness and substrate. Prepared granular quartz in the size range 4-250  $\mu\text{m}$ , packed in quartz equivalent containers with walls of sufficient thickness to provide secondary electron equilibrium, was irradiated with a known photon dose (3Gy). Each laboratory performed a calibration of their beta source using quartz of their preferred grain-size range (e.g. 90-

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<sup>2</sup> Roskilde, Denmark, DK 4000.

150  $\mu\text{m}$ ) taken from the irradiated material using a TL or OSL procedure, as described more fully by Göksu et al. (1995).

A combination of direct and indirect experimental methods was applied to determine  $D_{\text{BG}}$ . The indirect methods of high-resolution gamma-ray spectrometry (e.g. Murray et al. 1987) and thick source alpha counting (TSAC, see Aitken 1998) were employed to measure the concentrations of the natural radionuclides  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  (not measured by TSAC) in brick and uncontaminated soils, both of which were dried before measurement. The gamma-ray spectra were also measured to determine the extent of disequilibrium between the parents  $^{238}\text{U}$  and  $^{232}\text{Th}$  and detected progeny. The concentrations of anthropogenic  $^{137}\text{Cs}$  (together with the radionuclides of lithogenic origin,  $^{214}\text{Bi}$ ,  $^{214}\text{Pb}$ ,  $^{228}\text{Ac}$ ,  $^{212}\text{Pb}$ ,  $^{212}\text{Bi}$ ,  $^{208}\text{Tl}$  and  $^{40}\text{K}$ ) in air-dried soil samples were determined by gamma-ray spectrometry, performed with a high purity Ge detector of 15% efficiency (type 1GC1519<sup>3</sup>), shielded by 10 cm of low activity lead and coupled to a multichannel analyzer (type SNIP 204<sup>4</sup>). The gamma spectrometer had been calibrated by the Russian Bureau of Standards with an estimated accuracy of better than  $\pm 3\%$ . The values of specific activity for soil samples due to  $^{137}\text{Cs}$  derived from gamma spectrometry measurements are given in Table 3. The concentration values were converted to point-absorber infinite medium dose-rates in brick and soil using published conversion data (Adamiec and Aitken 1998).

The direct method of beta thermoluminescence dosimetry ( $\beta$ -TLD; Bailiff and Aitken 1980; Göksu and Bulur 1999) was applied to samples of crushed brick, yielding the point-absorber infinite-medium beta dose-rate within the sample measured. Luminescent dosimeters ( $\text{Al}_2\text{O}_3\text{:C}$  chips, 1 mm thick; Akselrod et al. 1990) were also deposited in walls at the sampled locations for  $\sim 1$  year to measure the average in-situ gamma and cosmic-ray dose-rate at a particular depth,  $\dot{D}_{\text{cap}}$ , due to

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<sup>3</sup> Manufactured by Detecktor Systeme GmbH, Germany.

natural sources of radiation. The laboratory beta source dose-rate for the 1 mm-thick  $\text{Al}_2\text{O}_3\text{:C}$  chips was determined separately following a similar calibration procedure to that discussed above for quartz grains, except that the known photon dose administered to the  $\text{Al}_2\text{O}_3\text{:C}$  chips by the  $^{60}\text{Co}$  source was significantly lower ( $<10$  mGy; Kalchgruber et al. 2002).

The gamma and cosmic components of the dose-rate are expected to vary with location in the wall, and consequently the value of  $D_{\text{BG}}$ , calculated using either Eq. 2 (Locs 71-74) or Eq. 3 (remaining locations), is for a specific position depth range within the wall. Although luminescent dosimeters were deposited at most of the locations discussed in this paper, dosimeter results were not obtained for Locations 71-74 due to either theft or destruction of the building.

## **Results**

### **Determination of $D_{\text{T}}$**

The average values of  $D_{\text{T}}$  for samples extracted from the depth range specified are listed in Table 2. The data have been grouped according to whether the presence of external sources of artificial radiation were detected experimentally (Locs 71, 73, 82/83) or not (the remainder). The  $D_{\text{T}}$  values represent the average value obtained by three measuring laboratories for the depth range indicated and the associated uncertainty given is the standard error of the mean value, given as a measure of precision. The distribution of  $D_{\text{T}}$  values was approximately normal for most samples and the number of determinations used to determine the mean value,  $n$ , is also indicated. It is worth noting that the dispersion of  $D_{\text{T}}$  values and the occurrence of outliers were generally higher than expected on the basis of work with bricks examined in the Chernobyl study (Bailiff et al. 2003). For the majority of samples the standard deviation associated with the determination of  $D_{\text{T}}$  by a single laboratory for a single depth range was less than  $\pm 15\%$ , but in two cases the datasets contained

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<sup>4</sup> Manufactured by Silena International Spa, Via Firenze, 3-20063 Cernusco sul Naviglio, Italy.

determinations with high standard deviations ( $\pm 40\%$ , 81-1 and  $\pm 27\%$ , 85-1). The behaviour associated with the latter was not always consistently found by all three laboratories for the same sample and an averaging of results produced acceptable precision. Although this behaviour may be partly attributed to the mineralogical composition of the sand added in the manufacture of the brick, a number of other causes connected with the dosimetry could account for the variability and requires further investigation.

### **Determination of $D_{BG}$**

The average value of  $D_{BG}$  for the appropriate depth range of each sample and the documented age of the building is given in Table 2, calculated using either Eq. 2 or Eq. 3 where the results of the dosimeter measurements were available. The beta dose-rate,  $\dot{D}_\beta$ , was obtained by calculating the average value of the results produced using the direct and indirect techniques discussed above and includes a reduction of the point-absorber dose-rates to account for attenuation effects due to the finite size of the quartz grains (Mejdahl 1979; 8-10% for the grain size ranges used in these experiments). For all locations including those where *in situ* dosimeter measurements were performed, the annual gamma dose-rate was calculated as a function of depth in the wall using a simplified model (Bailiff 2001; Bailiff et al. 2003). The geometry factors for gamma radiation emitted by radionuclides of lithogenic origin were calculated employing data derived from MC simulations by Loevborg (reproduced in Aitken 1985) from which the absorbed dose fractions were calculated for gamma radiation. The cosmic ray dose at sea level was estimated to be  $0.2 \text{ mGy a}^{-1}$  in the immediate sub-surface of the ground using data presented by Prescott and Hutton (1988). The value of  $\dot{D}_{\cos}$  was estimated to be  $0.15 \text{ mGy a}^{-1}$  at depths of between 5 and 120 mm from the wall surface and at the standard sampling height, making a nominal 30% allowance for attenuation due to the building structure. For those locations where dosimeter results were available, the measured dose-rate  $\dot{D}_{\text{cap}}$  was adjusted to obtain the combined gamma and cosmic-ray dose-rate for the

required depth range. Estimates of  $\dot{D}_\gamma$  calculated using data obtained by the direct (dosimeter) and indirect methods were compared and found to agree within 10%, indicating that the approximations used in the simplified model referred to above were reasonable.

### **Calculation of $D_X$**

The values of  $D_X$  given in Table 2 for each depth range(s) tested were calculated by substituting the relevant values of  $D_T$  and  $D_{BG}$  into Eq. 1. The overall error associated with  $D_X$  (68% level of confidence), was calculated by taking into account both random measurement errors and estimated systematic errors (Bailiff 1997; Bailiff et al. 2003).

### **Discussion and analysis**

Within the limits of experimental uncertainty no significant difference between values of  $D_T$  for slices at depths of ~10 and ~100 mm was found at Locations 72 (Akkol'), 74 (Bol'shaya Vladimirovka), 75 (Izvestka), 78 and 79 (Rubtsovsk), 80 and 81 (Laptev Log) and 85 (Kuria). Consequently the values of  $D_T$  given (Table 2) represent the average of the values of  $D_T$  obtained within the full depth range examined (5-100 mm). The absence of a significant external dose contribution in these cases is further supported by the calculated values of  $D_X$  (Table 2), the  $1\sigma$  ranges for which overlap with a value of 0. On the basis of the estimated uncertainties associated with the values of  $D_X$  the results obtained at these locations suggest that any cumulative dose due to artificial sources, if present, does not exceed ~25 mGy. On the basis of the experimental data for samples from Location 76 (Kainar), and taking into account that the levels of extant  $^{137}\text{Cs}$  concentration in soil (Table 3) are higher than at Dolon', we suspect that the bricks tested had either been manufactured after cessation of the major SNTS tests or had been subjected to repeated heating temperatures greater than 150 °C since construction of the chimney, and were therefore



unsuitable for retrospective dose measurements. We suspect the former to be correct since a reduction in  $D_T$  with depth (i.e. towards the inner part of the chimney) was not detected.

The values of  $D_T$  given in Table 2 for the external bricks correspond to sub-surface brick layers that are most appropriate in calculating the absorbed dose at the reference location,  $D_{RL}$ , and this is discussed further below. The values of  $D_X$  when plotted as a function of depth, provide a depth-dose profile, as shown in Fig. 5 for Loc. 71-2. The form of this profile provides experimental confirmation that the wall was exposed to external artificial sources of radiation (Meckbach et al. 1996; Bailiff 1999). Superimposed on the experimental data is a calculated profile obtained from MC simulations of external irradiation of the wall by radionuclide sources of energy 662 keV uniformly distributed on the ground to a depth of  $5 \text{ g cm}^{-2}$ . In this work we have made use of depth-dose and conversion factor calculations based on MC simulations performed in a previous study in the Chernobyl region (Jacob et al. 2000) where the major contributor to the absorbed dose was due to  $^{137}\text{Cs}$ . Consequently we have used depth-dose profiles for comparative purposes only and not to assign a particular time-averaged source energy. It should be noted that there is an unavoidable increase in the overall uncertainty associated with  $D_X$  where  $D_{BG}$  is a high proportion of  $D_T$ , particularly at greater depths in the brick (Bailiff 1997; ICRU 2002), as is the case for Loc. 71 where the building is almost 100 years old. The interpretation of the data obtained for each location is discussed individually.

### **Dolon'**

Two aspects of the construction of the building at Dolon' (Fig. 3a) are relevant to the interpretation of the results: i) as mentioned above, available evidence of the building history indicates that the original building had a broken façade and that the walls were covered with render of thickness 10-15 mm shortly after the construction of the buildings, and ii) a substantial portico was constructed

in ca 1952. A high proportion (~95%) of the external dose due to fallout from the 1949 test was delivered during the first year following the explosion (Shoikhet et al. 1998). Hence, for the purposes of this preliminary study, we assumed that the construction of the portico did not significantly affect the absorbed dose due to fallout from the 1949 test and that absorbed dose arising from extant fallout or fallout due to tests after 1952 was not significantly reduced. As indicated in Fig. 3a, Loc. 71-2 and Loc. 71-3 are spatially close but differ in terms of irradiation geometry since Loc. 71-3 is along the adjacent return wall orthogonal to the main face of the building.

The depth-dose profile obtained for Location 71-2 is shown in Fig. 5. The profile for 71-3 (not shown) is similar in form. The profile for the highly shielded interior sample (i.e. Loc. 71-4) was consistent with that calculated for radiation sources of lithogenic origin (i.e., natural), and the difference of  $16 \pm 29$  mGy between the average values of  $D_T$  and  $D_{BG}$  is considered not significant within the limits of experimental uncertainty.

The cumulative absorbed dose in air at the reference location,  $_{RL}D_X$  was obtained by using values of  $C_{RL}$  (Table 2) calculated for the sample depth range below the exposed surface (20-30 mm) at Loc. 71-2 ( $2.6 \pm 0.25$ ) and Loc. 71-3 ( $3.6 \pm 0.35$ ). On the basis of the depth-dose profile, the sources were assumed to have an average energy comparable to that for  $^{137}\text{Cs}$  (662 keV) and to have been uniformly distributed on the ground surface to a depth of  $5 \text{ g cm}^{-2}$ . This was considered to be a reasonable approximation in view of the correspondence between the calculated and experimental depth-dose profiles. Since there is little difference in the calculated conversion factors for ground and cloud sources for samples taken at ~1 m above ground level (ICRU 2002), the assumption that the sources were effectively ground-based is not expected to introduce additional uncertainty to the calculation of dose at the Reference Location.

Based on MC simulations for Location 71, the value for  $C_{RL}$  for location 71-3 was obtained by increasing the value of  $C_{RL}$  calculated for a standard plane wall geometry (Bailiff et al. 2003) by 30% (Fig. 3a) which is due to the additional shielding provided by the broken façade. No significant change in the value of the conversion factor is predicted by calculation associated with the difference in sampling height (~1 m at Loc.71-2 vs ~2 m at Loc. 71-3). Because of the relatively short half-lives of the isotopes present in the fallout, any evidence of the spatial variation of radionuclide distribution is not available. As discussed above we assumed that a high proportion of the dose due to fallout had been delivered before the portico and large concrete terrace were added. If this assumption were incorrect we would expect the value of  $D_X$  at Loc. 71-3 to be a substantially higher fraction of that obtained at Loc. 71-2. In the absence of information to the contrary, uniformity of deposition was assumed and no adjustment for either local heterogeneity or the addition of shielding was attempted. Allowances made for a significant increase in the concentration of fallout near to the walls of the building would result in a lowering of the conversion factor, and *vice versa* (Bailiff et al. 2003). An allowance for these uncertainties was made when estimating the uncertainties assigned to the values of  $C_{RL}$ . Based on the data given in Tables 1 and 2 and the assumptions discussed above, the estimates obtained for the cumulative absorbed dose in air at the reference location,  $_{RL}D_X$ , are  $475 \pm 110$  mGy and  $415 \pm 140$  mGy for Locations 71-2 and 71-3, respectively.

A further aspect of the experimental results should be noted. If the radionuclide sources were contained in a semi-infinite cloud, the values of  $D_X$  at Locs 71-2 and 71-3 would be expected to be roughly equivalent. Although the precision obtained in  $D_X$  is not sufficient to draw firm conclusions, the nominal agreement between the calculated (70%) and experimental (67%) ratios for dose in brick at the two locations consequently provides evidence that there is no indication that

the dose should be attributed to a cloud source. Although this is not a critical issue for calculating dose at the reference location, it has potentially important implications in dose reconstruction.

### **Kanonerka**

The house sampled in Kanonerka is distinguished from the other buildings in the study by having walls constructed of brick on the ground floor on which was mounted a wooden superstructure (Fig. 3c). It is likely that a balcony extending across the front and rear of the house, now removed, had been originally present at the time of the tests. Of the three samples taken, two were from external walls (73-1, facing the main street and 73-2, at the rear) and the other (73-3) was from a heavily shielded location within the interior of the house. The depth-dose profile for 73-1 (Fig. 6) indicates the presence of external artificial sources of radiation, and the profile obtained for Loc. 73-2 is similar in form but has a less pronounced slope at greater depths in the brick. The profile for the shielded interior sample (Loc. 73-3) was consistent with that calculated for radiation sources of lithogenic origin, and the difference of  $32 \pm 26$  mGy between the average values of  $D_T$  and  $D_{BG}$  indicates that assessment of the latter is satisfactory.

The depth-dose profiles obtained for Loc. 73-1 and Loc. 73-2 are more complex to interpret than those obtained in Dolon'. At depths greater than 20 mm the profile for Loc. 73-1 (Fig. 6) broadly corresponds to the calculated profile for a ground-based source of energy 662 keV. However, some contradictory results were obtained for depths less than 20 mm where a subset of the results indicate significantly higher values of  $D_X$  and a slope that is significantly steeper than at greater depths. The concordance of results obtained within each subset using both TL and OSL procedures suggests that experimental dose evaluation procedure is not the main cause of the differences.

If the subsets of data points are measures of absorbed dose due to external sources, the steeper component of the profile has a 'half depth' of between 5 and 10 mm. This could result from the presence of short-lived sources of significantly lower photon energy (x or gamma emitting isotopes) and/or bremsstrahlung arising from beta particles adhering to the front surface of the brick. Fallout collected on the balcony and washed off onto the wall surface below during cleaning could cause a highly heterogeneous distribution of dose in the sub-surface layers across the brick (e.g. via cracks in the surface layers of the brick), leading to different values of dose according to the section of brick examined, noting here that each laboratory obtained a 'set' of results with a different section of brick. In addition it should be noted that since the mass energy absorption coefficient for quartz increases substantially for energies below 100 keV (ICRU 1992), the fraction of the absorbed dose due to low energy photons would be overestimated using the experimental procedure applied in this study since the laboratory beta sources were calibrated against a high energy photon source ( $^{60}\text{Co}$ ). Another possibility is that mortar containing quartz grains contributing strong luminescent signals (associated with a geological dose) could have become incorporated with the brick sample during the initial preparation treatment. Although the sample preparation procedures were designed to avoid this possibility, we cannot rule out its occurrence. We concluded that in either case the data sub-set with higher values of dose in the sub-surface region should be excluded and that the calculation of dose in air at the reference location should be based on the absorbed dose determinations due to the more highly penetrating gamma radiation. An estimate of  $225 \pm 60$  mGy was obtained for  $_{\text{RL}}D_X$  based on the value of  $D_X$  ( $125 \pm 30$  mGy) obtained after exclusion of the upper component of the profile and a value of  $1.8 \pm 0.2$  estimated for  $C_{\text{RL}}$ .

At the rear of the house (Loc. 73-2), the experimental and calculated profiles ( $E=662$  keV) were generally consistent in form, but for each set of data (i.e. obtained by each laboratory) the experimental values diverge from the calculated profile, lying above the latter at depths  $>40$  mm.

Although a calculated profile resembling the experimental profile can be obtained by assuming that some of the sources were located on the interior surface of a cavity in the wall, underestimation of  $D_{BG}$  also causes divergence of a generally similar type. However, the uncertainty in the experimental profile at greater depths (e.g. >60 mm) is not sufficient to distinguish between the two possible causes. Our calculations indicate that at Loc. 73-2 the contribution to the absorbed dose measured in the outer layers of the sampled brick would not be significantly affected by the presence of the sources located on an inner wall surface. Since the value of  $D_X$  calculated for the interior shielded sample (Loc. 73-3) is  $32 \pm 26$  mGy, the possibility of underestimation of  $D_{BG}$  is not excluded, but in view of the levels of uncertainty associated with the  $D_X$  we did not attempt to adjust the value of  $D_{BG}$ . An estimate of  $250 \pm 60$  mGy was obtained for  $_{RL}D_X$  at this location based on values of  $140 \pm 30$  mGy and  $1.8 \pm 0.2$  for  $D_X$  and  $C_{RL}$ , respectively. Assuming that both sampled walls (Locs 73-1 and 73-2) were facing similarly contaminated ground, the weighted average of the values of  $D_X$  for both locations is  $133 \pm 28$  mGy and, using a value for  $C_{RL}$  of  $1.8 \pm 0.2$ , the estimated value of  $_{RL}D_X$  at Loc. 73 is  $240 \pm 60$  mGy.

### **Leshoz Topolinskiy**

The height of the 4-story building in Leshoz Topolinskiy enabled samples at ground level (Loc. 82-1) and at ~12 m elevation (Loc. 83-1) to be obtained. This is of interest because comparison of the results can potentially yield information concerning the time-averaged source configuration. Although the accessible part of the building at ground level suffered some fire damage several years ago, it was considered unlikely that the external bricks in the 90 cm thick wall were heated during the fire to temperatures sufficient to cause thermal fading of the latent luminescence signal. The depth-dose profile obtained for Loc. 83-1 (Fig. 7) confirms the presence of external artificial sources of radiation, and the profile for Loc. 82-1 (not shown) is qualitatively similar in form. Although there is overall agreement between different laboratories within experimental error (95%

level of confidence), there were general difficulties in obtaining high levels of precision in  $D_T$  for sample from depths between 20 and 100 mm, for the reasons mentioned above. However, comparison of the value of  $D_T$  (290 mGy) for a heavily shielded sample (235 mm depth) with the calculated value of  $D_{BG}$  at the same depth (270 mGy) indicates that the determinations are reliable.

The experimental depth-dose profile for Loc. 83-1 (not shown) is broadly consistent with the calculated depth-dose profile for a ground source (to  $5 \text{ g cm}^{-2}$ ) of energy 662 keV. Although full MC simulations have not been completed, initial calculations indicate that the profile calculated for a section of wall at elevated height is slightly steeper than that for ground level samples. The average values of  $D_X$  for Locations 82-1 (1 m) and 83-1 (12 m), calculated for the 3-10 mm depth range, are  $126 \pm 37$  mGy and  $90 \pm 27$  mGy, respectively (Table 2).

As the height above ground level increases, the value of  $C_{RL}$  changes and, moreover, its value is more sensitive to source configuration (e.g. ground-based vs cloud) at elevated height compared with samples at 1 m. Relevant calculations based on MC simulations have been performed by Meckbach (in Jacob et al. 2000) for samples at elevations of 1 m and 10 m in a building for the same or similarly contaminated land. The absorbed dose in brick at 10 m elevation is predicted to be 81% of the value at 1 m if the fallout has penetrated the ground (i.e. to  $5 \text{ g cm}^{-2}$ ), whereas if the fallout remains on the surface, the absorbed dose at 10 m is predicted to drop to 57 % of the value at 1 m. If, on the other hand, the fallout is contained entirely in a cloud source, the absorbed dose in brick is expected to be 22% higher at 10 m than that at 1 m. For Locs 83 and 82, the experimentally determined value of the ratio  $_{12m}D_X/_{1m}D_X$  is  $0.7 \pm 0.1(5)$ . If we assume that results for 10 m and 12 m are equivalent, this ratio corresponds more closely with the time-averaged configuration of sources that are contained in the sub-surface of the ground (0.7-0.9) rather than a cloud source (1.2). Although the interpretation is inevitably limited given the experimental uncertainty achieved, it

illustrates the potential value of samples at elevated heights that can provide further information concerning source distribution where the extant fallout activity is not detectable. Based on this information, and using the value of  $D_X$  obtained for Location 82 and  $C_{RL}$  values for ground sources with a depth of penetration of fallout ( $5 \text{ g cm}^{-2}$ ), the resulting value of  ${}_{RL}D_X$  is  $230 \pm 70 \text{ mGy}$ .

### **Comparison of luminescence and calculated estimates**

The luminescence estimates of the cumulative absorbed dose in air due to gamma radiation at the Reference Location can be compared with previously published calculated dose estimates for settlements in Table 4. The published values of dose derived from exposure rate data obtained after the nuclear test were extracted from sources for Kazakhstan and Altai (Logachev 1997; Shoikhet et al. 1998) that are based on the same archived primary data. We considered two issues when attempting to compare previously published absorbed dose estimates based on dose-rate measurements performed after a short delay following the tests with those produced by retrospective luminescence dosimetry:

- i) the lack of detailed dosimetry data for settlements;
- ii) the possible strong heterogeneity in the fallout pattern, in particular that due to the 1949 test.

The combination of these two factors currently limits the opportunity to derive cumulative estimates of gamma dose for the sampled locality derived from modelling calculations for comparison with the luminescence results. However there is an opportunity to extrapolate the published calculated estimates in Dolon' because of the particular circumstances of the fallout and the history of previous dosimetry measurements.

On the basis of measurements of wind speed on 29 August 1949 of about  $60 \text{ km h}^{-1}$  (Shoikhet et al. 1998) and the distance to ground zero (118 km; Logachev 2002) the radioactive cloud is estimated



to have reached Dolon' within about 2 hours. Also during the day of the test it rained intermittently. Aerial radiation surveys moving in a north easterly direction from the SNTS were performed on 5 September and ground surveys during the period 7-13 September. These measurements showed a rise in gamma dose-rate at a distance of 100-110 km NE of the test site (Shoikhet et al. 1998), i.e. in the vicinity of Dolon'. Since the wind speed was relatively high, the absorbed dose due to radiation from the cloud is likely to have been small compared with that due to fallout deposited on the ground (also supported by the experimental evidence). From maps published by Logachev (1997) the nearest ground dose-rate measurements appear to have been performed in 1949 about 5 km to the south-west of the village and, according to Shoikhet et al. (1998), the pattern of the fallout deposition near Dolon' village was estimated to comprise a narrow 2 km corridor of maximum dose rate, decreasing by a factor of 4 at a further distance 3-4 km. Later measurements (Tsyb et al. 1990, Logachev 2002 and this work) of  $^{137}\text{Cs}$  activity in soil within the settlement and its environs support this overall picture. Logachev (2002) reported that moving from the SSE region to the NNW region of the settlement, covering about 3.5-4 km across the settlement towards the central axis of the plume (located about 1.5 km from the northern perimeter of the settlement), the  $^{137}\text{Cs}$  areal activity increased by a factor of 14.5 from  $0.74 \text{ kBq m}^{-2}$  to  $10.7 \text{ kBq m}^{-2}$ . Although the exact locations of these measurements are not clear, the evidence overall indicates that Dolon' was not uniformly irradiated due to a heterogeneous distribution of fallout across the settlement and we suspect that a comprehensive survey of extant  $^{137}\text{Cs}$  would confirm a relatively narrow plume of fallout within the settlement. From measurements of  $^{137}\text{Cs}$  in undisturbed soil 2 km to the NW of Dolon', designated by SNTS specialists in 1989 as the "maximum contaminated spot near the village" (Tsyb et al. 1990), the areal activity due to  $^{137}\text{Cs}$  was estimated to be  $8.9 \text{ kBq m}^{-2}$  (Stepanenko et al. 1989; Tsyb et al. 1990). Published estimates of mean external dose for Dolon' village (Table 4) of  $\sim 1100 \text{ mGy}$  (Stepanenko et al. 1994),  $\sim 1500 \text{ mGy}$  (Stepanov et al. 2002) and  $\sim 130 \text{ R}$  (Logachev 1997) were obtained were based on the maximum value of measured dose-rates obtained from the archives,

later published by Logachev (1997) and Shoikhet et al. (1998). Since the former church is located in the S of Dolon', an estimate of the mean external dose for this part of the settlement was derived from the estimated range of 1100-1500 mGy by applying a scaling factor of 0.42, which yields a range of 460-630 mGy. The scaling factor corresponds to the ratio of the  $^{137}\text{Cs}$  areal activities for soil from S Dolon' ( $3.74 \text{ kBq m}^{-2}$ , adjusted to 1989) and NW Dolon' ( $8.9 \text{ kBq m}^{-2}$ ), where the former was derived from the measured specific activity of soil taken from the forest along the southern perimeter of the settlement (Table 3), and where the conversion assumes a core weight of 1.46 kg and cross-sectional area of  $0.015 \text{ m}^2$ . It should be noted that these comparisons are based on soil contamination measurements performed at *one* location only on each occasion (near the village in 1989 and within it in 1999). The calculated estimate compares well with the luminescence determination for  $_{\text{RL}}D_X$  of  $475 \pm 110 \text{ mGy}$ . Given the approximations made and the complexity of the dosimetry this agreement may be judged to be fortuitous, but we consider it to be a very promising outcome given the exploratory nature of the study. Our estimate of dose in air for the church location in Dolon' is lower than that ( $\sim 1.4 \text{ Gy}$  in air) reported by Takada et al. (2002). To obtain their estimate Takada and co-workers applied a conversion factor from dose in ceramic to dose in air for a plane wall geometry, using a value of 2. If the sources were predominantly ground-based this value would not be appropriate for the corner location (Fig. 3a), from where the samples were taken (M. Hoshi, University of Hiroshima, personal communication)

The luminescence results for locations in Akkol', Bol'shaya Vladimirovka, and Izvestka in Kazakhstan and Rubtsovsk and Kuria, selected as settlements where the dose due to fallout was expected to be comparatively low, are consistent with the published calculated data. At these locations the dose due to fallout could not be distinguished from the cumulative natural background dose within a margin that we estimate to be  $\pm 25 \text{ mGy}$ . At Kainar it was not possible to obtain a conclusive result and we suspect that the bricks tested were manufactured after the tests. Given that

the levels of extant  $^{137}\text{Cs}$  in soil were higher than those found at Dolon', further investigation of this settlement is needed. In contrast, in Kanonerka, where the predicted cumulative dose was about 15% of that in Dolon', the luminescence results indicate a cumulative dose that is significantly higher (~60%) than the published calculated dose. Although there are caveats associated with the experimental results for Kanonerka, they are sufficient to indicate that a closer inspection of this settlement is needed. Of the remaining locations in Altai the outcomes of comparisons are mixed. At Laptev Log a dose due to fallout could not be detected (i.e.,  $< 25$  mGy), contrasting with the published calculated value of 480 mSv. At Kuria the experimental results suggest no detected fallout dose ( $D_X = 7 \pm 28$  mGy) whereas the calculated dose is 43 mSv. At Leshoz Topolinskiy, the luminescence estimate of the cumulative dose ( $230 \pm 70$  mGy) is about one sixth of the dose predicted by calculation (1400 mSv; Shoikhet et al. 1998) for the settlement. In both Laptev Log and Leshoz Topolinskiy heterogeneity in fallout distribution may account for these differences, both underlining the potential difficulties of accurate dose reconstruction in settlements and the necessity of performing an investigation of contemporary  $^{137}\text{Cs}$  areal activity in the vicinity of each (luminescence) sampling location, and also more widely within the settlement.

In the absence of suitable  $^{137}\text{Cs}$  activity data for soil samples that were directly associated with the calculated estimates of dose for settlements we have not attempted to scale our results for specific activity (Table 3) at sampled locations, other than at Dolon'. However, some observations can be made concerning the activity data for the remaining locations, particularly since they provide some evidence of the delivery of fallout to ground level. On the basis of our activity data for Dolon' ( $\sim 31$  kBq  $\text{kg}^{-1}$ ) and assuming that  $^{137}\text{Cs}$  activity is an accurate proxy for total fallout deposition, we would expect to detect a dose due to fallout for locations where the activity exceeded  $\sim 4$  kBq  $\text{kg}^{-1}$ . This calculation is based on a minimum detectable value of 25 mGy for  $D_X$  that corresponds to about 15% of  $D_X$  measured at Loc. 71-2. In the cases of Laptev Log, Rubstovsk (Loc.78) and

Bol'shaya Vladimirovskaya the absence of a detectable fallout dose is consistent with the low levels of extant  $^{137}\text{Cs}$  in the soil sub-surface. There are three locations that are counter to this finding, where significant  $^{137}\text{Cs}$  activity was measured but where no fallout dose was detected. They are at the second location in Rubtsovsk (Loc. 79;  $\sim 15 \text{ kBq kg}^{-1}$ ), at Kuria ( $14 \text{ Bq kg}^{-1}$ ) and at Izvestka ( $26 \text{ Bq kg}^{-1}$ ). The abnormal activity profile at Rubtsovsk (Loc. 79, activity  $>20 \text{ cm}$ ) may explain the failure to detect a fallout dose, but at Kuria there is no obvious explanation, and in both cases the relatively greater distances from the NTS could be a relevant factor. The riverine location of the kiln at Izvestka could have led to the progressive deposition of  $^{137}\text{Cs}$  carried within fluvial sediments and deposited within the flood plain. As discussed above, the bricks from Kainar appear to have been manufactured after the tests, but the high levels of  $^{137}\text{Cs}$  concentration in soils from the village and the surrounding area underline the need for further investigation. At the remaining locations (Dolon', Leshoz Topolinskiy and Kanonerka) the significantly higher levels of  $^{137}\text{Cs}$  are concordant with elevated values of  $D_X$  but there is an absence of a simple proportionality.

In summary, the majority of published dose estimates for this region are based on a very limited quantity of dose rate and radionuclide concentration measurements following the delivery of fallout. The locations and points of available archive data regarding dose rate measurements in September 1949 do not coincide with the settlements. Additional comparisons of luminescence and calculated estimates of dose require further measurements of extant  $^{137}\text{Cs}$  concentration with depth in soil within and outside settlements to test the degree of heterogeneity. Additionally, searches for the existence of  $^{137}\text{Cs}$  measurements performed within the region in 1949 may yield further data.

## Conclusions

Settlements within the regions of highest predicted dose and those of lowest predicted dose within the lateral tail of the published fallout trace map due to the August 1949 tests were examined. Of

the ten settlements where brick samples were taken, luminescence estimates of cumulative absorbed dose in air due to fallout at the reference location were obtained for Dolon' and Kanonerka in Kazakhstan and Leshoz Topolinskiy in Altai (Russia). These values, in excess of the cumulative natural background, are  $475\pm110$  mGy,  $240\pm60$  mGy and  $230\pm70$  mGy respectively. The dose estimates derived from the luminescence results for the locality of the former church in Dolon' village are in agreement with published calculated estimates of dose normalized to  $^{137}\text{Cs}$  concentration in soil. It is also interesting to note that an estimate of cumulative dose of less than 500 mGy has been obtained based on a study of ten inhabitants of Dolon' village using biological dosimetry techniques (Salomaa et al. 2002). The experimental results obtained at Dolon' provided the potentially interesting indication that radiation from sources on the ground rather than in a cloud was dominant in contributing to the fallout dose,  $D_X$ . These results also illustrate the importance of building geometry when selecting samples and on the other the opportunity to use differences in shielding as a tool in investigating source geometry.

Comparison of luminescence and calculated estimates of cumulative dose for Kanonerka and Leshoz Topolinskiy will require further computation by modelling specialists. Estimates of cumulative absorbed dose for samples from the remaining settlements (Akkol', Bol'shaya Vladimrovka and Izvestka in Kazakhstan, and Rubtsovsk, Laptev Log and Kuria in Altai, Russia) yielded values of cumulative absorbed dose that indicated no significant dose due to fallout that could be detected within a margin of about 25 mGy.

Although the cumulative dose at Laptev Log was expected to be about half the published value for Dolon', heterogeneity of fallout distribution may account for the apparent discrepancy. This provides further evidence that in general a high degree of heterogeneity in the distribution of fallout is likely to have occurred within the area of the plume. Significant variations in radionuclide

concentrations in soils have been observed in settlements. This may be the combined result of a relatively narrow radioactive trace and unstable weather conditions during the delivery of fallout. Where published mean dose estimates are based on extrapolated data derived from sporadic monitoring, significant differences between estimates of cumulative dose produced by the two methods are likely to occur. Consequently it is important that any future work addresses the problem of mapping such variations in order to provide the basis for accurate dose reconstruction in settlements. The relatively narrow corridor of fallout distribution due to the 29 August 1949 nuclear test in the region investigated amplifies this problem.

This study has shown that method of luminescence retrospective dosimetry has the potential to provide estimates of cumulative radiation dose in contaminated populated areas that can be applied to dose reconstruction studies for this region. In particular, providing suitable buildings can be found, it has the potential to be applied to both investigate the nature of the heterogeneity in the distribution of fallout associated with the 1949 test at Semipalatinsk and to provide independent values of cumulative gamma dose that can be compared with those produced by calculation .

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## **The application of retrospective luminescence dosimetry in areas affected by fallout from the Semipalatinsk Nuclear Test Site: an evaluation of potential**

### **Figure Captions**

**Figure 1.** Regional map showing the relationship between the SNTS, major cities and sampled settlements. Kainar is outside the region shown. The locations of the sampled settlements are identified in Table 1. The dose contours associated with the plume from the 1949 test are based on calculations by Shoikhet et al. (1998). The contour values correspond to the following levels of cumulative dose: 1 (250 mSv); 2 (50 mSv); 3 (10 mSv); 4 (1 mSv).

**Figure 2.** Calculated depth-dose profiles at an height of 1 m above ground level in a brick wall exposed to gamma radiation originating from radionuclides deposited on the ground, for source energies of 140 keV (open diamonds) and 662 keV (open triangles). Shown for comparison is the calculated depth-dose profile where the radionuclides are distributed on the surface of the wall and the source energy is 662 keV. The depth-dose profiles have been normalized to a layer of depth 1 cm from the surface of the brick.

**Figure 3 a-e.** Photographs of sampled buildings at Dolon', Kanonerka, and Leshoz Topolinskiy: a) the former church in Dolon' (Loc. 71) showing the broken façade, classical portico and concrete terrace added after the 1949 tests; b) the extraction of cores from the front face of the broken façade, Loc. 71-2, as shown; Loc. 71-3 is just above the limit of the photograph on the return wall. The excavated area at the corner is presumed to be the location of samples taken by the University of Hiroshima in ca 1995; c) the abandoned merchant's house in Kanonerka (Loc. 73) showing the wooden superstructure, where the balcony, now removed, was located at a level corresponding to the top of the brick walls; d) a close up of Loc. 73 indicating a the extraction of a whole brick and a core hole to the right); e) the substantial mill at Leshoz Topolinsky showing the west facing wall (gable end; Loc. 82) and south facing walls; Loc. 83 is located just below the level of the roof on the north facing wall (hidden from view).

**Figure 4.** Schematic diagram showing dissection and coding of whole bricks. In the example shown the brick, taken from Loc. 'n', is divided into four slices. The code for the first slice is 'n'-1-1. If a

slice is divided a further identification number is allocated, in this case the code for the right half of the divided slice 4 is 'n'-1-4-2.

**Figure 5.** *Comparison of experimental and calculated relative depth-dose profile for cores from Dolon', Loc. 71-2. The experimental results obtained using TL and OSL procedures, by the participating laboratories, as indicated, represent the net dose due to fallout,  $D_x$ , after subtraction of the cumulative natural background dose. For one sample (71-2-3) the MRRC laboratory sent extracted quartz to the DUR and GSF laboratories for evaluation. Results labelled MRRC-HEL refer to sample prepared in the MRRC laboratory and measured in the HEL laboratory by staff from both laboratories. The solid line is an interpolated curve fitted to the calculated values (Calc GS) obtained from MC simulations assuming a uniform distribution of radionuclides soil with average energy 662 keV to a depth of  $5 \text{ g cm}^{-2}$ .*

**Figure 6.** Comparison of experimental and calculated relative depth-dose profile for cores from Kanonerka, Loc. 73-1. Experimental and computational details as described in the caption to Fig.5.

**Figure 7.** Comparison of experimental and calculated relative depth-dose profile for cores from Leshoz Topolinskiy, Loc. 83. Experimental and computational details as described in the caption to Fig.5.

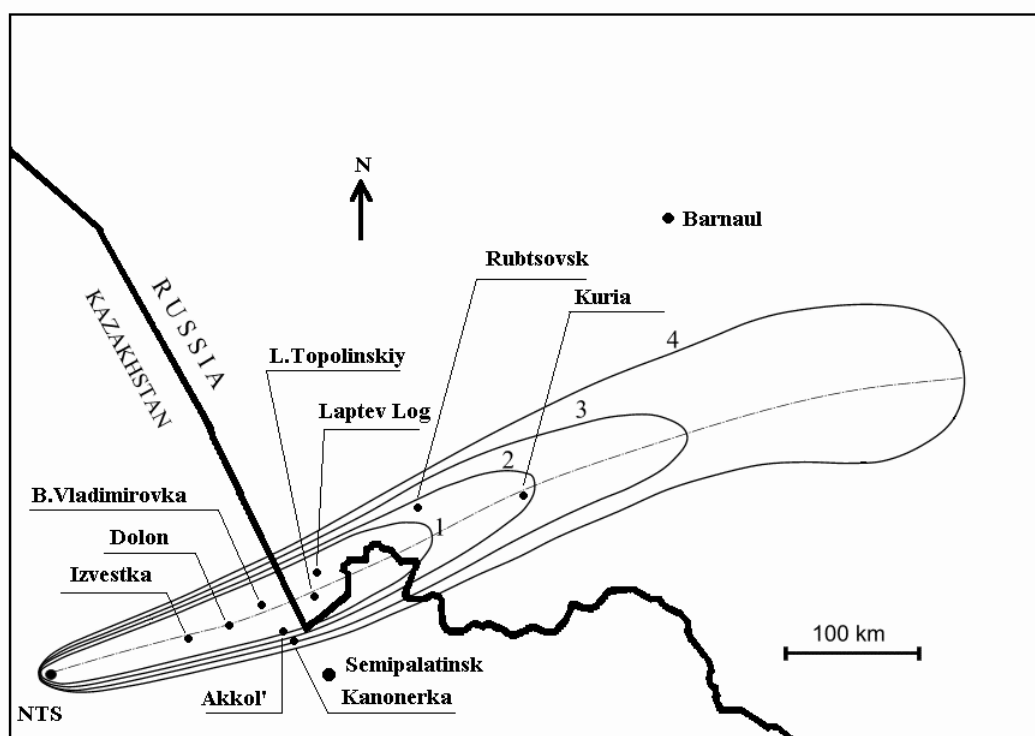


Fig. 1



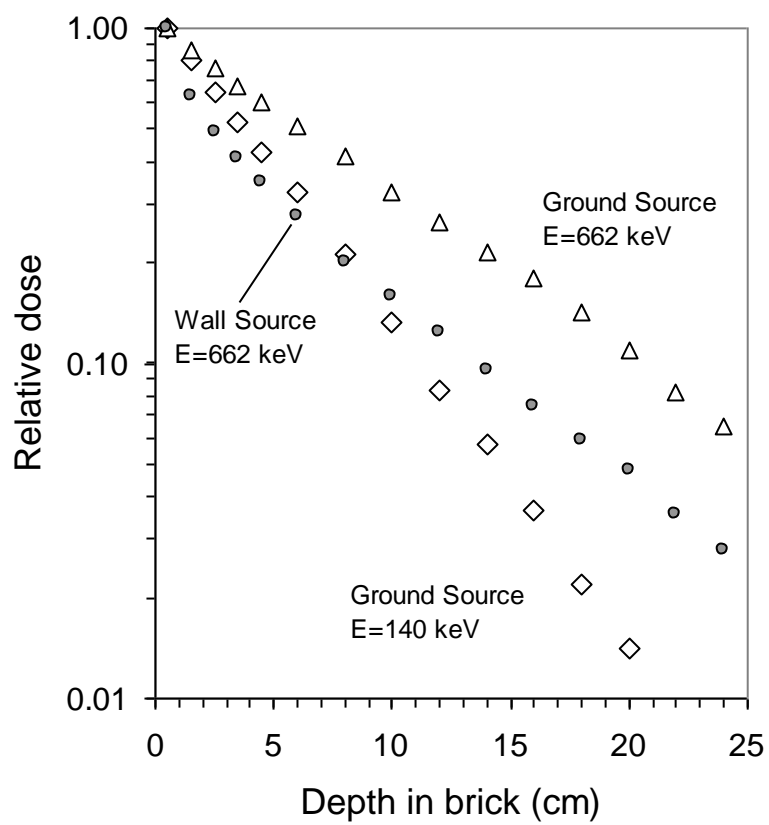


Fig. 2



a)



b)

Fig. 3



c)



d)

Fig. 3



e)

Fig. 3

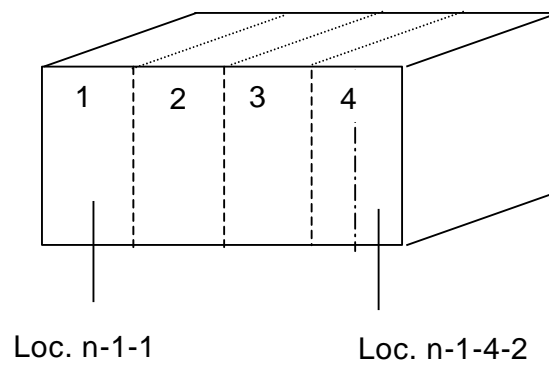


Fig. 4.

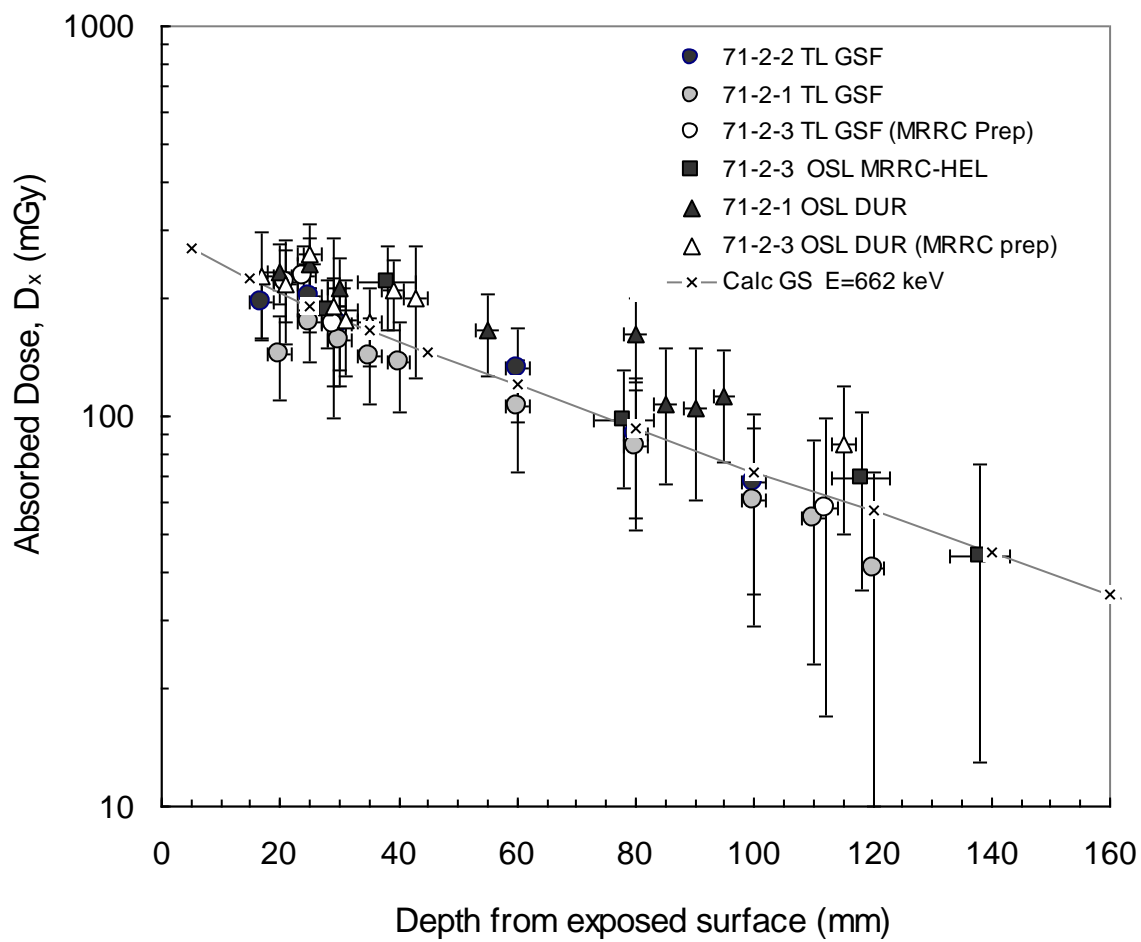


Fig. 5

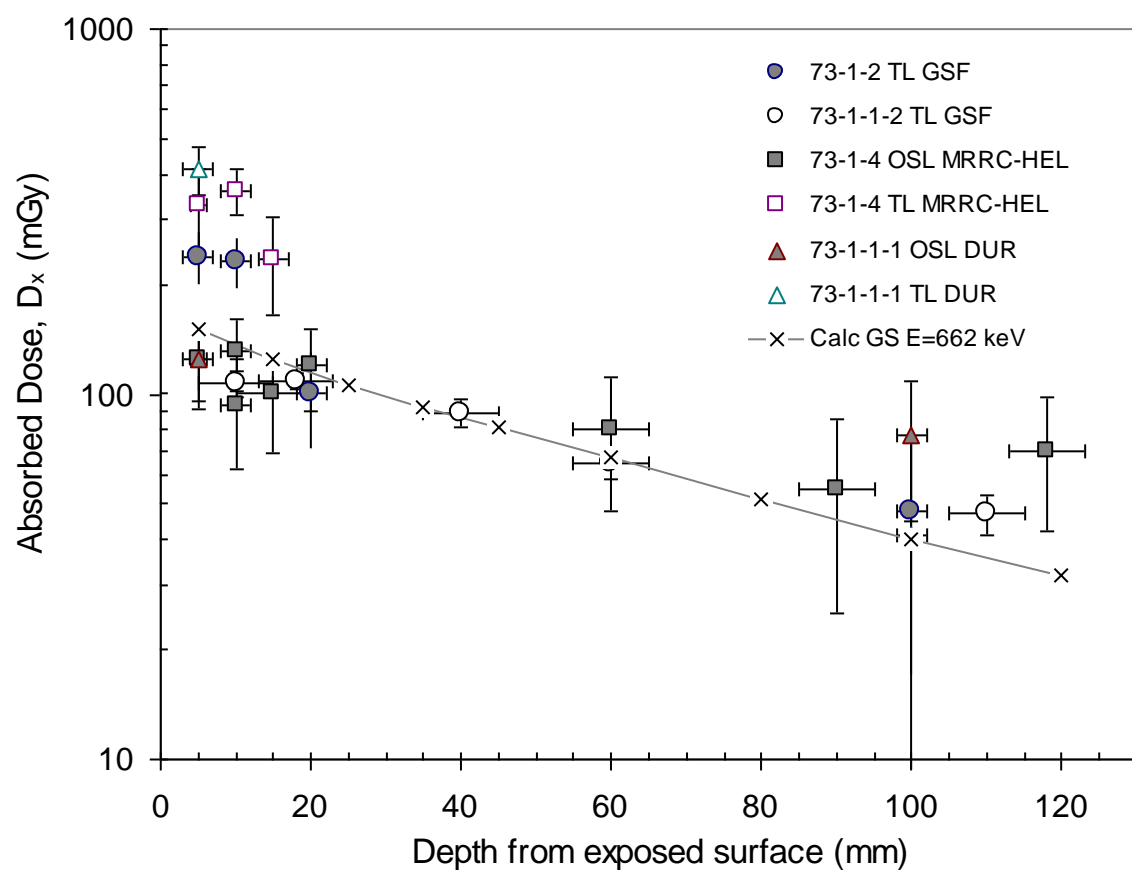


Fig. 6

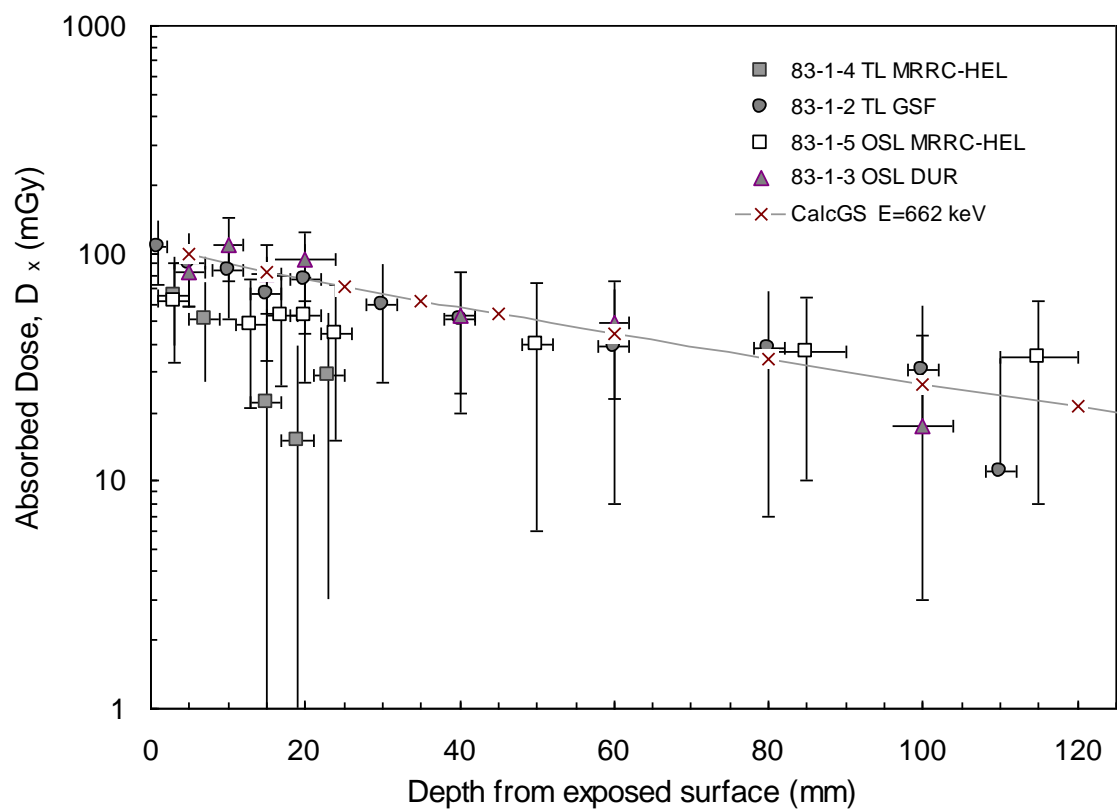


Fig. 7



# The application of retrospective luminescence dosimetry in areas affected by fallout from the Semipalatinsk Nuclear Test Site: an evaluation of potential

## Tables

**Table 1.** A summary of sampled building locations, their documented or reported age and the orientation of the sampled area given in terms of the cardinal points for external samples. Samples from interior shielded locations are indicated as 'int'. The map locations refer to the numbered locations marked on the regional map shown in Fig. 1. The ages of the buildings correspond to the difference between the recorded/reported date of construction of the building and date when samples were taken in Kazakhstan (1999) and in Altai (2000). It has been assumed that the bricks were manufactured shortly before use.

Settlement	Building	Location	Age (y)	Location #	Orientation	Height of sample (m)	Comments
<i>Kazakhstan</i>							
Dolon'	Former Church	N50°39'54" E 79°18'42"	95±2	71-2 71-3 71-4	S W Int	1.2 2.9 1.2	~15 mm render
Akkol'	Timber Mosque	N 50°40'49" E 79°49'57"	>70	72	S-E	0.8	Brick foundations
Kanonerka	House	N 50°43'44" E 79°41'07"	87±2	73-1 73-2 73-3	W E Int	2.0 2.0	
Bol'shaya Vladimirovka	House	N 50°53'11" E 79°29'10"	80±5	74-1 74-2 74-3	N-E S-W Int	0.8 1.1 1.0	~10 mm render
Izvestka	Disused lime kiln	N 50°37'48" E 78°51'30"	80±4	75-1 75-2 75-3	S-W S-W E	1.4 3.0 1.8	
Kainar	Chimney, adobe house	N 49°11'57" E 77°23'05"	>50	76-1	N	3.0	
<i>Altai</i>							
Rubtsovsk	House	N 51°30'27" E 81°12'15"	73±2	78-1	S-W	1.0	
Rubtsovsk	Former barracks	N 51°31'39" E 81°11'36"	53±2	79-1	S-E	1.0	
Laptev Log	Tractor workshop	N 51°04'12" E 80°12'05"	66±4	80-1 81-1	W E	1.3 1.3	
Leshoz	Mill	N 50°56'31"	90±2	82-1	W	1.4	Four-story
Topolonskiy		E 80°04'40"		83-1	N	13.0	
Kuria	Former church	N 51°36'02" E 82°17'15"	98±2	85-1 86-1	W W	1.5 6.4	Two-story

**Table 2.** Summary of dosimetry results for each external (ext) or internal (int) wall location, including values of key parameters in the determination of  ${}_{RL}D_x$ . The uncertainties are standard errors of the unweighted mean values (68% level of confidence). Those associated with  $D_T$  and  $D_{BG}$  are due to random sources, and those associated with  $D_x$  and  ${}_{RL}D_x$  include an assessment of both random and systematic sources. The values given in the seventh column (n) correspond to the number of determinations of  $D_T$ . The conversion factor  $C_{RL}$  for Loc. 71-3 differs from that for Loc. 71-2 due to additional shielding, as discussed in the main text. The calculation of  $D_x$  at Kainar (Loc. 76-1) is discussed in the main text.

Location	Wall	Code	Depth (mm)	Avg. dose rate @1m ( $\mu\text{Sv}\cdot\text{h}^{-1}$ )	$D_T$ (mGy)	n	$D_{BG}$ (mGy)	$D_x$ (mGy)	$C_{RL}$	${}_{RL}D_x$ (mGy)
Dolon'	ext	71-2	20-30	0.08	508 $\pm$ 11	8	326 $\pm$ 13	182 $\pm$ 38	2.6 $\pm$ 0.2(5)	475 $\pm$ 110
	ext	71-3	20-30	0.10	462 $\pm$ 18	4	340 $\pm$ 13	122 $\pm$ 39	3.4 $\pm$ 0.3(5)	415 $\pm$ 140
	int	71-4	20-120	0.12	351 $\pm$ 16	5	335 $\pm$ 13	16 $\pm$ 29	-	-
Kanonerka: House, front	ext	73-1	3-10	0.08	401 $\pm$ 15	5	276 $\pm$ 11	125 $\pm$ 30	1.8 $\pm$ 0.2	225 $\pm$ 60
Kanonerka: House, rear	ext	73-2	3-10	0.10	387 $\pm$ 10	5	247 $\pm$ 11	140 $\pm$ 30	1.8 $\pm$ 0.2	250 $\pm$ 60
	int	73-3	5-100	0.09	289 $\pm$ 10	3	257 $\pm$ 12	32 $\pm$ 26	-	-
Leshoz	ext	82-1	3-10	0.06	400 $\pm$ 23	7	274 $\pm$ 11	126 $\pm$ 37	1.8 $\pm$ 0.2	230 $\pm$ 70
Topolinskiy: Mill, lower										
Mill, upper	ext	83-1	3-10	0.11	335 $\pm$ 9	5	245 $\pm$ 11	90 $\pm$ 27	-	-
Akkol'	ext	72-1	5-100	0.07	242 $\pm$ 11	13	240 $\pm$ 18	2 $\pm$ 28		
Bol'shaya Vladimirovka	ext	74-2	5-100	0.09	258 $\pm$ 2	4	263 $\pm$ 17	-5 $\pm$ 26		
Izvestka	ext	75-1	5-100	0.11	265 $\pm$ 16	14	263 $\pm$ 16	2 $\pm$ 28		
Kainar	ext	76-1	5-100	0.16	82 $\pm$ 4	11	209 $\pm$ 8	-		
Rubtsovsk	ext	78-1	5-100	0.11	216 $\pm$ 6	11	230 $\pm$ 10	-14 $\pm$ 21		
Rubtsovsk	ext	79-1	5-100	0.07	182 $\pm$ 17	6	170 $\pm$ 9	12 $\pm$ 23		
Laptev Log	ext	80-1	5-100	0.07	211 $\pm$ 4	15	211 $\pm$ 10	0 $\pm$ 19		
Laptev Log	ext	81-1	5-100	0.08	181 $\pm$ 15	9	198 $\pm$ 10	-17 $\pm$ 23		
Kuria	ext	85-1	5-100	0.10	317 $\pm$ 8	7	310 $\pm$ 13	7 $\pm$ 28		

**Table 3.**  $^{137}\text{Cs}$  concentration data given as specific activity ( $\text{Bq kg}^{-1}$ ) for soils sampled at the depths indicated and measured in 1999 (Kazakhstan) and in 2000 (Altai) in the vicinity of sampled settlements.

Settlement	Location(s)	Depth	<sup>137</sup> Cs
			Activity
		(cm)	Bq kg <sup>-1</sup>
<i>Kazakhstan</i>			
Dolon'	Near Forest	0-5	30.5±2.7
		5-20	≤0.1
Akkol'	72	0-20	7.3±0.7
Kanonerka	Adj. village	0-20	17.9±1.7
B. Vladimirovka	74	0-20	<0.2
Izvestka	150 m from	0-5	25.6±2.3
	Loc. 75	5-10	1.1±0.2
		10-100	<1
Kainar	Village	0-5	10.4±1.0
		5-10	20.7±1.9
		10-15	14.5±1.3
		15-40	≤1
	10 km NW	0-3	78±6.9
	of village	3-6	5.2±0.6
		6-40	<1
<i>Altai</i>			
Rubtsovsk	78	0-5	1.1±0.3
		5-10	3.2±0.4
		10-15	3.5±0.4
		15-20	4.0±0.5
		20-25	1.8±0.3
		25-30	5.0±0.5
Rubtsovsk	79	0-5	3.1±0.4
		5-20	<1
		20-25	12.9±1.2
		25-30	18.5±1.7
		30-35	14.7±1.4
		35-40	16.3±1.6
Laptev Log	80/81	0-5	4.1±0.5
		5-10	<1
		10-15	3.7±0.4
		15-40	<1
Leshoz	82/83	0-5	35.8±3.2
Topolinskiy		5-10	7.3±0.7
		10-40	<1
Kuria	85	0-5	13.9±1.3
		5-10	10±1
		10-15	8.1±0.8
		15-20	8.1±0.8
		20-25	4.8±0.5
		25-30	1.0±0.2

**Table 4.** Comparison of  $_{RL}D_x$  and available published calculated cumulative external dose (given in dose units as published) for settlements in Kazakhstan and Altai, respectively) for measurement points closest to the settlements from which brick samples were obtained. The published dose is based on available results of dose-rate and radionuclide measurements following the nuclear test, where the source is given in the adjacent column (1, Stepanov et al. 2002; 2, Stepanenko et al. 1994; 3, Logachev 1997; 4, Shoikhet et al. 1998). The distances between the sampled settlement and the nearest point of dose-rate measurement performed in September 1949 are based on an examination of schematic maps published by Logachev (1997; 2002) and Shoikhet et al. (1998). R is an historical unit of exposure in air; to convert to exposure in SI units ( $C\ kg^{-1}$ ) multiply the value shown by  $2.58\ 10^{-4}$  ( $C\ kg^{-1}$  per R); 1 R is numerically equal to about 8.7 mGy in air (STP) and for gamma photons  $> 100\ keV$ .

Settlement	Location(s)	$_{RL}D_x$ (mGy)	Published Dose	Source	Distance to settlement (km)
<i>Kazakhstan</i>					
Dolon'	71-2	475±110	900 mSv 1100 mGy 130 R	1 2 3	5-8
Akkol'	71-3	415±140			
Kanonerka	72	<25			
	73-1	225±60	15 R	3	3-5
	73-2	250±60			
Bol'shaya Vladimirovka	74	<25	<1 R	3	
Izvestka	75	<25	<0.1 R	3	10-15
Kainar	76	-	9 R	3	
<i>Altai</i>					
Rubtsovsk	78	<25	(mSv) 30	4	2-3
Rubtsovsk	79	<25	30	"	2-3
Laptev Log	80, 81	<25	480	"	3-5
Leshoz Topolonskiy	82-1	230±70	1400	"	5-7
Kuria	85, 86	<25	43	"	1-2